Diverging length scale and upper critical dimension in the Mode-Coupling Theory of the glass transition

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Abstract. – We show that the glass transition predicted by the Mode-Coupling Theory (MCT) is a critical phenomenon with a diverging length and time scale associated to the cooperativity of the dynamics. We obtain the scaling exponents ν and z that relate space and time scales to the distance from criticality, as well as the scaling form of the critical four-point correlation function. However, both these predictions and other well known MCT results are *mean-field* in nature and are thus expected to change below the upper critical dimension $d_c = 6$, as suggested by different forms of the Ginzburg criterion.

One of the most striking property of glassy materials is the extremely fast rise of their relaxation time (or viscosity) as the temperature is lowered or the density increased [1]. The basic mechanism for this spectacular slowing down is not well understood, but it is reasonable to think that it is intimately related to cooperative effects. The dynamics becomes sluggish because larger and larger regions of the material have to move simultaneously to allow for a substantial motion of individual particles. Although this qualitative idea has pervaded the glass literature for many years [1], it is only quite recently that a proper measure of cooperativity (and of the size of the rearranging regions) was proposed and measured experimentally [2] and in numerical simulations [3–5]. The idea is to measure how the 'unlocking' events are correlated in space; technically, this involves a four-point density correlation function (see below) from which one can extract a growing dynamical correlation length [4–6]. Recent extensive numerical simulations in Lennard-Jones systems have confirmed the crucial importance of this growing length scale for the dynamics of the system [4–6]. Furthermore, the four-point correlation function is found to have scaling properties similar to those expected near a critical point [4, 5, 7], suggesting that the physics of the glass transition should be understood as a critical phenomenon dominated by large scale fluctuations. This ingredient in fact appears in various forms in several recent phenomenological approaches [7–10]. This is, at first sight, in plain contradiction with the Mode-Coupling Theory (MCT) of the glass transition. MCT is considered by many to be the closest to a first principle, microscopic theory of glasses yet

achieved, with many qualitative and quantitative successes in explaining various experimental and numerical results [11–13]. However, freezing in MCT was argued repeatedly by its founders to be a small scale phenomenon, the self-consistent blocking of the particles in their local cages. Since no small wavevector singularities seem to exist in MCT, power-laws and scaling are only expected in time but not in space [12]. This is surprising since on general grounds a diverging relaxation time can only arise from processes involving an infinite number of particles. This point of view was challenged by Franz and Parisi [14] in the context of the so-called 'schematic' MCT (see also [8] for an early insight). This simplified version of the theory is (formally) equivalent to describing mean-field spin glasses with three body interactions, for which the physics is known in great details. At the (ergodicity breaking) critical temperature T_c, the curvature of the relevant TAP states is known to vanish [15]. Therefore, one expects, and indeed finds, that a susceptibility diverges when $T \to T_c$. This susceptibility turns out to be the precise analogue, in the context of this spin model, of the four-point correlation function mentioned above. By analogy with usual second order phase transitions, the analysis of [8, 14] suggests that the MCT freezing transition is in fact accompanied by the divergence of the correlation length of the four-point correlation function.

In this letter we show that the MCT dynamical transition in finite dimensions must indeed be understood as a critical phenomenon: dynamical correlations become long-range both *in time and space*, in agreement with the insight of [8,14]. We obtain the MCT dynamical scaling exponents ν and z that relate space and time scales to $|T - T_c|$, as well as the scaling form of the critical four-point correlation function. However, these results (as well as all other quantitative MCT predictions) are *mean field* in nature and change in dimensions less than $d_c = 6$ due to long-wavelength fluctuations, as confirmed by different forms of the Ginzburg criterion. Our strategy is similar to the one used for ordinary critical phenomena. Consider the ferromagnetic Ising transition as an example. In that case one can show that there exists a certain functional of the magnetisation field such that (a) its first derivative leads to exact equations for the magnetization and (b) its second derivative is the inverse of the spin-spin correlation function. In general one cannot compute this functional exactly but one can guess its form using symmetry arguments, or compute it approximately in a diagrammatic expansion. Its simplest version corresponds to the Ginzburg-Landau free energy functional. The saddle point equations for the magnetization then leads to the mean field description of the transition. One finds in particular that the singular behaviour at the transition is related to the vanishing of a 'mass'. This has two important implications: (1) the spin susceptibility diverges at the transition, (2) because of the vanishing mass the corrections to the mean field result computed by adding more diagrams blow up whenever $d < d_c = 4$, i.e. spatial fluctuations change the critical behavior. We shall show that exactly the same scenario takes place within the MCT of the glass transition, except that the order parameter is now a two point function, the dynamical density-density correlation function. The analogue of the spin-spin correlation function is therefore the following four point correlation:

$$G_4(\vec{r},t;\vec{\delta},\tau) = \langle\rho(0,0)\rho(\vec{\delta},\tau)\rho(\vec{r},t)\rho(\vec{r}+\vec{\delta},t+\tau)\rangle - \langle\rho(0,0)\rho(\vec{\delta},\tau)\rangle\langle\rho(\vec{r},t)\rho(\vec{r}+\vec{\delta},t+\tau)\rangle, \quad (1)$$

where $\rho(\vec{x},t)$ is the density fluctuation at position $\vec{x}$ and time t and $\langle\cdot\rangle$ denotes an average over the dynamics. Its intuitive interpretation (for example in the case where $\vec{\delta} = 0$) is as follows: if at point 0 an event has occurred that leads to a decorrelation of the local density over the time scale τ, what is the probability that a similar event has occurred a distance $\vec{r}$ away, within the same time interval τ, but shifted by t? In other words, $G_4(\vec{r},t;\vec{\delta},\tau)$ measures the cooperativity of the dynamics. Different values of $\vec{r}$ and t allow one to measure the full space-time structure of dynamical cooperativity, and are the analogue of the space and time separation entering the

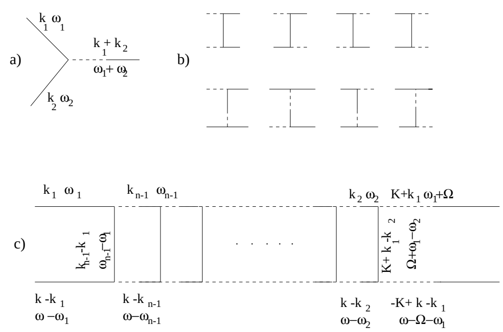

Fig. 1 – Basic vertex of the MCT (a), and a ladder diagram that contribute to the divergence of the four-point susceptibility (c). Other ladders can be obtained by combining the eight elementary blocks shown in (b). The full lines represent correlation functions, whereas the mixed dashed-full lines are response functions.

standard spin-spin correlation function in critical dynamics. The quantities $\vec{\delta}$ and τ, on the other hand, serve to define the 'order parameter', i.e. the density-density correlation function $C(\vec{\delta},\tau) = \langle \rho(0,0)\rho(\vec{\delta},\tau)\rangle$. Previous studies have focused on the case $t = 0$, and δ smaller than the particle radius, to which one can associate a 'susceptibility' $\chi_4(\tau)$ by integrating over space. More generally, we shall focus on a wave-vector dependent, a.c. susceptibility as:

$$\chi_4(\vec{k},\omega;\vec{K},\Omega) = \int d^d\vec{r}\, d^d\vec{\delta}\, dt\, d\tau\, e^{-i\vec{k}\cdot\vec{r}-i\omega t-i\vec{K}\cdot\vec{\delta}-i\Omega\tau}\, G_4(\vec{r},t;\vec{\delta},\tau). \tag{2}$$

The quantities G_4 or χ_4 can be computed as in the ferromagnetic case inverting the second derivative of an appropriate functional. Within any field theoretical derivation of MCT (e.g. the Das and Mazenko formulation [16] or more heuristic derivations [13, 17]), the functional alluded to above can be constructed as [18]:

$$F(G) = -\frac{1}{2}\mathrm{Tr}\log G + \frac{1}{2}\mathrm{Tr}\, G_0^{-1}G + \Phi_{2PI}(G) \tag{3}$$

where G and G_0 are compact notations for the full and the bare propagator of the theory, and $\Phi_{2PI}(G)$ is the sum of all two particle irreducible Feynman diagrams (that cannot be decomposed in two disjoint pieces by cutting two lines) constructed with the vertices of the theory and using the full propagator as line. The first derivative of F gives back the exact equations on G, whereas the four point function is obtained inverting its second derivative, which is nothing else than $G^{-1}G^{-1}$ (coming from the derivative of the first term in (3)) minus the derivative of the self-energy with respect to the two point function. Thus, since MCT is tantamount to only retaining the 'bubble' diagram, the four point functions are obtained from ladder diagrams, such as the one drawn in Fig. 1([1]).

In order to understand the mechanism that leads to a divergence of the four-point correlation, consider the ladder diagram shown in Fig. 1. The n^{th} order contribution to $\chi_4(\vec{k},\omega,\vec{K},\Omega)$ reads:

$$\Delta_n\chi_4(\vec{k},\omega,\vec{K},\Omega) = \int d^d\vec{k}_1 d^d\vec{k}_2 ... d^d\vec{k}_{n-1} \int d\omega_1 d\omega_2 ... d\omega_{n-1} C(\vec{k}_1,\omega_1)C(\vec{k}-\vec{k}_1,\omega-\omega_1) \times$$
$$\mathcal{M}_{\vec{k},\omega}(\vec{k}_1,\vec{k}_{n-1};\omega_1,\omega_{n-1})\mathcal{M}_{\vec{k},\omega}(\vec{k}_{n-1},\vec{k}_{n-2};\omega_{n-1},\omega_{n-2}) .... \mathcal{M}_{\vec{k},\omega}(\vec{k}_2,\vec{k}_1+\vec{K};\omega_2,\omega_1+\Omega)$$

([1])From a more general point of view, one can obtain all the MCT predictions using a Landau expansion of $F(G)$ in $G(t,T) - G(t=\infty,T_c)$, justified in dimension larger than six. This derivation, which unveils the generality of the MCT predictions, will the subject of a future publication.

where the matrix $\mathcal{M}$ is defined as:

$$\mathcal{M}_{\vec{k},\omega}(\vec{k}_2,\vec{k}_1;\omega_2,\omega_1) = T^2 \frac{R(\vec{k}_1,\omega_1)}{\rho k_1^2} \frac{R(\vec{k}-\vec{k}_1,\omega-\omega_1)}{\rho(k-k_1)^2} C(\vec{k}_2-\vec{k}_1,\omega_2-\omega_1) V(\vec{k}_1,\vec{k}_2,\vec{k}),$$

where ρ is the particle density and C is the correlation function and R the response function (assumed to be related to C by the fluctuation-dissipation theorem (2)) for the density in Fourier space, and

$$V = \{\vec{k}_1 \cdot [\vec{k}_2 c(\vec{k}_2) + (\vec{k}_1-\vec{k}_2)c(\vec{k}_1-\vec{k}_2)]\}\{(\vec{k}_1-\vec{k}) \cdot [(\vec{k}_2-\vec{k})c(\vec{k}_2-\vec{k}) + (\vec{k}_1-\vec{k}_2)c(\vec{k}_1-\vec{k}_2)]\}$$

where $c(\vec{k})$ is the direct correlation function. If one follows the so-called schematic approximation [11, 12] where all structure factors $S(\vec{k})$ appearing inside integrals are approximated by $A\delta(k-k_0)$ and all ingoing and outgoing momenta have modulus k_0, then all k-dependence disappears. Replacing the resulting (k-independent) four leg vertex $T^2\hat{V} = T^2 S(k_0)k_0 A^2/(4\pi^2\rho)$ with $T^2\hat{V}_{3-spin} = 3$ (where T is the temperature), one gets exactly the same diagrams of the mean field $p = 3$ case. For $T < T_c$ and for small frequencies, the correlation function acquires a (non-ergodic) contribution: $C(\omega) = f\delta(\omega) + C_{\rm reg}(\omega)$ (where f is usually denoted q in the spin-glass literature), while the response function tends to a constant at low frequencies given by $(1-f)/T$ (with power-law corrections in ω, see below). Therefore, one obtains, as the dominant contribution: $\Delta_n\chi_4(\omega,\Omega) = C(\Omega)\delta(\omega)f[\hat{V}(1-f)^2 f]^{(n-1)}$. Clearly, the series diverges when $(1-f)^2 f\hat{V} = 1$, an equation precisely satisfied at the mode coupling temperature T_c, and signaling criticality (see, e.g. [11, 12, 15]). Hence, the asymptotic value of $\chi_4(\tau\to\infty)$ is divergent at T_c. One can show, using a transfer matrix method to analyze all diagrams generated from the eight building blocks drawn in Fig. 1, that the above singularity is unchanged, i.e. χ_4 indeed diverges as $(1-(1-f)^2 f\hat{V})^{-1}$ (3). Since $f - f_c \sim \sqrt{\epsilon}$ for $T < T_c$, we conclude that χ_4 diverges as $\epsilon^{-1/2}$ for $T \nearrow T_c^-$, indeed in agreement with the results of [14].

Our aim in the following is to understand in details how this divergence is affected by non zero frequencies and wave-vectors. In order to do so, we rely on the detailed results known about C and R in the vicinity of T_c. One knows for example that when $T = T_c - \epsilon$, the plateau in C is reached after a time $\tau_f \sim \epsilon^{-1/2a}$, where a is a non-trivial exponent. In the regime $\tau_f^{-1} \ll \omega \ll 1$, the response function acquires an extra contribution proportional to ω^a [11]. On the other hand, when $T = T_c + \epsilon$, two time scales diverge with different exponents. One is again the time τ_f to reach the (pseudo-)plateau value f of the correlation function, and the second is the *terminal* time $\tau_t \sim \epsilon^{-\gamma}$ (with $\gamma = 1/2a + 1/2b$), beyond which the correlation finally drops to zero. Both the exponents a and b are non universal; for example, in the mean-field $p = 3$ case one has $a \approx 0.395$ and $b = 1$. Using these results for $T < T_c$ and for non zero ω (corresponding to the shift in the measurement times for the four-point correlation function) we find that the term $1-(1-f)^2 f\hat{V} \sim \sqrt{\epsilon}$ leading to the singularity, is replaced by $\sqrt{\epsilon} + Z\omega^a$, where Z is a constant. Thus, the only characteristic time scale is, for $T < T_c$, the plateau time τ_f. Resumming the series $\sum_n \Delta_n\chi_4$ is much more subtle in the case $T > T_c$. A naive transposition of the results for $T < T_c$ to $T > T_c$ suggests the following: since the $1/\sqrt{\epsilon}$ divergence of χ_4 for $T < T_c$ can be traced back to $f_c - f \sim \sqrt{\epsilon}$, then the divergence for $T > T_c$ should be $1/\epsilon$ because now $f - f_c \simeq \epsilon$. Furthermore, from the form of the ladder diagrams one expects that both the correlation time scale in the t direction and the peak time

(2)This property is certainly true for the $p = 3$ spin-glass for $T > T_c$ but may be problematic for MCT. However, we believe that the following conclusions are independent of the strict validity of FDT.

(3)Actually a complete calculation should also take into account the diagrams with extra vertices coming from imposing an initial condition at equilibrium. Although these diagrams do not change the singular behavior for $\tau \to \infty$ they are responsible for the vanishing of χ_4 when $t \to \infty$.

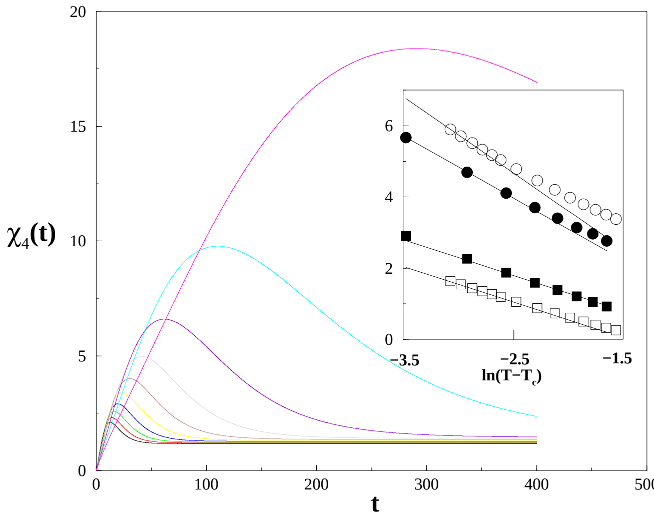

Fig. 2 – Four-point susceptibility $\chi_4(\tau)$ in the mean-field $p = 3$ spin-glass, obtained by integrating the dynamical equations obtained in [14], for various temperatures $T > T_c$ ($T_c = 0.612$). The inset shows, in log-log scale, the position t^* (black circles) and the height $\chi_4(t^*)$ (black squares) of the peak as a function of $T - T_c$, and the plain lines are power-laws with exponents -1 and $-(1 + a)/2a = -1.76$, corresponding to the 'naive' argument discussed in the text. We confirmed these results by also solving numerically the mixed $p = 3 + 6$ case, for which $a = 0.345$ and $b = 0.717$ (open symbols). These last curves have been shifted for clarity.

t^* of $\chi_4(\tau)$ are given by the terminal time scale τ_t. The above predictions turn out to be in perfect agreement with the numerical solution of the $p = 3$ mean-field case shown in Fig. 2 ([4]), and can in fact be obtained analytically using another route [19].

Returning to MCT in finite dimensions, one has to include the wavevector dependence of C, R and V. Assuming ergodicity is broken, χ_4 for $\vec{k} = 0$ is found to diverge when the largest eigenvalue of matrix M is equal to unity:

$$M(\vec{k}_2, \vec{k}_1) = \frac{\rho}{k_2^4} S(\vec{k}_1) S(\vec{k}_2) S(\vec{k}_1 - \vec{k}_2) V(\vec{k}_1, \vec{k}_2, \vec{0})(1 - f_{k_1})^2 f_{\vec{k}_2 - \vec{k}_1} \tag{4}$$

with, for $T \leq T_c$: $f_q/(1 - f_q) = \frac{\rho}{2q^4} \int d^3k'/(2\pi)^3 S(\vec{q}) S(\vec{k}') S(\vec{q} - \vec{k}') V(\vec{q}, \vec{k}', \vec{0}) f_k f_{\vec{q}-\vec{k}'}$. The spectrum of M has been previously studied in detail because it controls the convergence of the above equation on f_q when solved by an iterative procedure. It has been established (see [12]) that the maximum eigenvalue of M is non degenerate and approaches one as $\sqrt{\epsilon}$ at the transition, which in turn leads to the famous MCT singularity of $f - f_c$. The effect of a non zero wave-vector $\vec{k}$ can be understood using perturbation theory. By symmetry, it is clear that the correction to the largest eigenvalue must be, for small $\vec{k}$, of order k^2. This leads for small k and $T < T_c$ to a propagator $(\Gamma k^2 + \sqrt{\epsilon})^{-1}$. Naively transposing the above results for $T > T_c$ gives a propagator behaving as $(\Gamma' k^2 + \epsilon)^{-1}$. Thus, we conclude that the four-point correlation computed, say, for $t = 0$ and $\vec{\delta}$ of the order of the particle radius, and τ of the order of the correlation time scale (τ_f for $T < T_c$ and τ_t for $T > T_c$) will behave as the two-point

([4])Note that these numerical results do not coincide with the results reported in [14], obtained with the same code; in particular, the height of the peak was found to diverge as $\epsilon^{-1/2}$ and not as ϵ^{-1}. A possibility is that the 'conjugated field' used in [14] was not small enough.

correlation in standard critical phenomena, i.e as $\mathcal{G}\left(\frac{r}{\ell}\right)/r^{d-2+\eta}$. From the above results, we find $\eta = 0$, and a dynamical length ℓ that diverges as $\epsilon^{-\nu}$, with $\nu = 1/4$, $1/2$ respectively below and above the transition. The above scaling law for $T > T_c$ is compatible with recent numerical simulations [5–7]. It is also interesting to note that for $T = T_c$ and for small k, ω our results imply that χ_4^{-1} behaves as $\Gamma k^2 + Z\omega^a$, which allows us to identify the dynamical exponent as $z = 2/a$. At the critical point, length and time scales are related by an anomalous sub-diffusion exponent: $r \sim t^{a/2}$, with $a < 1/2$ [12]. The above result will hold for $T > T_c$ and $\tau_f^{-1} \ll \omega \ll 1$, whereas for $\omega\tau_t \ll 1$ the denominator of the propagator will rather behave as $k^4 + (\tau_t\omega)^2$, corresponding to simple diffusion on long time scales. Note that the relation between ℓ and τ_t allows one to define a second dynamical exponent $z' = 1/a + 1/b = 2\gamma$, echoing the presence of two diverging time scales for $T > T_c$. Numerically, for the Kob-Andersen LJ mixture, $z' = 4.5 \pm 0.2$ [7], a value surprisingly close to our prediction $2\gamma = 4.68$ [12]. However, exactly as for usual critical phenomena, one should expect long-wavelength fluctuations to be dominant below some upper critical dimension d_c and changing the value of all the exponents, *at least sufficiently close to* T_c. The value of d_c can be obtained by analyzing the diagrams correcting the MCT contribution to the self-energy considered here, or using a Ginzburg-like criterion; both lead to $d_c = 6$. The diagrammatic is in fact very similar to that of the ϕ^3 theory, which is in a sense expected since the order parameter is here the correlation function itself, which does not have the Ising symmetry. More physically, one can argue that for the spatial fluctuations to be irrelevant, these should not blur the $\sqrt{\epsilon}$ singularity for $T < T_c$ (or the ϵ singularity for $T > T_c$) of the non-ergodic parameter f around f_c. Within a sphere of radius ℓ, these fluctuations are of order $\left[\ell^d \int^\ell r^{d-1} G_4(r) dr\right]^{1/2} \sim \ell^{(d+2)/2}$, to be compared to the total contribution of the singularity, $\ell^d\sqrt{\epsilon}$ for $T < T_c$ (or $\ell^d\epsilon$ for $T > T_c$). Using $\epsilon \sim \ell^{-4}$ for $T < T_c$ ($\epsilon \sim \ell^{-2}$ for $T > T_c$), we thus find that the fluctuations become dominant for large ℓ whenever $(d+2)/2 > d-2$, or $d < d_c = 6$. A detailed calculation of the exponents for $d < 6$ would be very interesting, to estimate how the mean field exponents of MCT are affected by spatial fluctuations in $d = 3$. A naive guess, based on a percolation interpretation of the MCT transition [20], suggests $\nu \approx 0.88$ and $\eta \approx 0$. [The analogy with phase transitions could also shed light on the non trivial (fractal) structure of the mobile regions, see [21–23].] The knowledge of these critical exponents is crucial, since a quantitative fit of many experimental results has been attempted using MCT [12]. In particular, neither the $\sqrt{\epsilon}$ singularity of the non-ergodic parameter for $T < T_c$ (argued to be a signature of the MCT singularity) nor the exponents derived above for χ_4 are expected to remain valid for $d = 3$. Nevertheless the existence of a diverging length scale could hint at a large (and perhaps unexpected) degree of universality in the dynamics of glassy systems [5, 7].
The critical fluctuations discussed above should however not be confused with another type of fluctuations that are expected to destroy the MCT singularity altogether, and suppress the glass transition. These fluctuations are the 'activated events' or 'hopping processes' that prevent a complete freezing of the super-cooled liquid, but the consistent inclusion of these in an extended version of MCT is still quite a challenge [12, 16]. The way the two types of fluctuations interact to make any of the above finite dimensional MCT predictions observable is unclear to us: a second Ginzburg criterion, pertaining to activated fluctuations, should be devised and compared to the one above.

The existence of a diverging dynamical correlation length puts strong constraints on any theory of glass forming liquids. We have shown that finite dimensional MCT does, at least qualitatively, survive the test (note that the existence of a diverging length scale in MCT should allow for the observed decoupling between diffusion and viscosity.) Only a more quantitative comparison of the predictions of MCT in three dimensions would allow one to rule it

out entirely, or to confirm that it is indeed a useful picture (at least sufficiently far from T_c). Simulations of systems with different fragilities and/or in higher dimensions would also be very interesting to gain further insight, and test different predictions or scenarii. For instance, the critical mobility defect scenario of [7] predicts an upper critical dimension $d_c = 4$ with the same dynamical exponent $z \approx 3.7$ for both strong and fragile-to-strong liquids, in contrast with the prediction of spatial MCT, where $d_c = 6$ and where both a and b (and therefore z) are system-dependent. Finally, the extension of the ideas proposed here to non-zero shear rates and aging situations would be extremely valuable (see [23, 24]).

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